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Cold anti-Stokes photoluminescence of InP self-assembled quantum dots in the presence of electric current

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Abstract. An intense anti-Stokes photoluminescence is observed in a structure with InP quantum dots in the presence of a direct electric current and cw optical pumping below the lowest electron hole transition in the quantum dots. A simple model is proposed for the explanation of the observed phenomenon. Its essential point is the existence of deep level defects around the QDs.

Introduction

A cold anti-Stokes photoluminescence (AS PL) has been observed in various semiconductor heterostructures [1–5]. Usualy Auger and two-step excitation mechanisms are discussed to be responsible for this effect. On the other hand, there is a well known electroluminescence (EL) effect which occurs when an electric current flows through a specially designed heterostructure like laser diode. In the present work, we show that in structures with InP quantum dots (QDs) it is possible to observe a strong AS PL at low temperature when the optical pumping and electric current are applied to the structure simultaneously. There is no AS PL if one of the actions is absent.

1 Experimental

We studied the heterostructure schematically drawn in the inset of the Fig. 1(a). It was grown by gas source molecular beam epitaxy on a n^+ GaAs substrate. The QDs were formed by the deposition of InP on the InGaP layer and covered by the top InGaP layer. The areal density of QDs is about 10^{10} cm^{-2}. The average base diameter is $\approx$ 50 nm and the height is $\approx$ 10 nm. The studied sample was supplied with a semitransparent gold Shottky contact on the top surface and an ohmic contact on the back surface.

The PL and PL excitation (PLE) spectra were recorded by using the setup including a double monochromator U1000, a cw Ti:sapphire laser and a photon counting system with a cooled GaAs photomultiplier tube. All the measurements were done at the sample temperature of 5 K.

2 Experimental results

In Fig. 1(a) the PL spectra of InP QDs are shown. They were recorded at resonant (a) and nonresonant (b) laser excitation with constant intensity and different electric currents I in the range of 0–1000 μA/mm^2. It is found that under positive $U_{\text{bias}} > 0.7$ V ("+" on the top of the sample) the electric current through the sample increases rapidly. AS PL appears synchronously with the electric current and its intensity also increases rapidly with the bias. At $I = 1000\,\mu$A/mm^2, the integral intensity of AS PL under resonant excitation (Fig. 1(a)) exceeds that of the Stokes PL without the electric current. This means that the observed

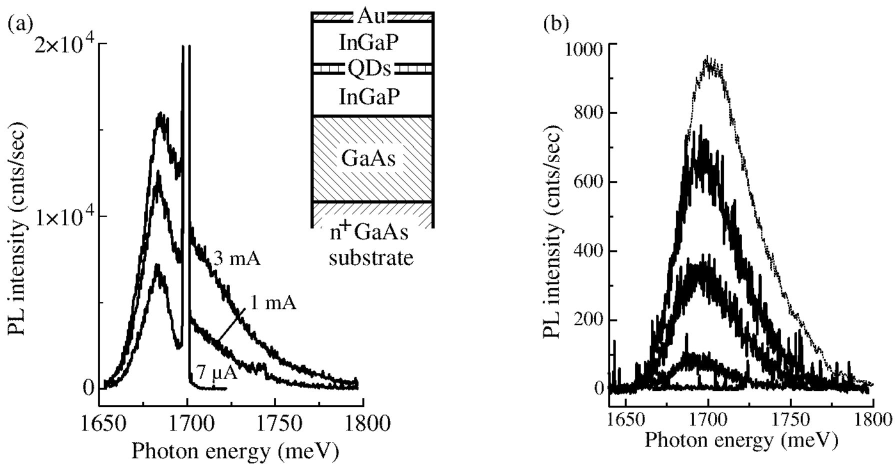

Fig. 1. (a) Stokes and anti-Stokes PL of InP QDs under resonant excitation in the presence of an electric current through the heterostructure (excitation photon energy $E_{\rm exc} = 1700$ meV and excitation power $P_{\rm exc} = 100$ W/cm^2). Inset: A simplified structure of the studied sample.
(b) AS PL at the low energy excitation ($E_{\rm exc} = 1494$ meV, $P_{\rm exc} = 500$ W/cm^2) under different electric currents trough the sample (from the bottom to the top: 130 μA, 290 μA, 1030 μA, and 3300 μA). The sample area is $S = 3$ mm^2. The Stokes PL spectrum of QDs under the high photon energy excitation ($E_{\rm exc} = 1800$ meV) is shown by a dotted line for comparison.

effect is very strong. Under negative bias, the AS PL is not observed and the Stokes PL decreases [6].

AS PL is observed under the optical excitation with photon energy $E_{\rm exc}$ much lower than the energy of photons emitted by QDs. The PL excitation (PLE) spectrum of the sample is shown in Fig. 2. It should be stressed that AS PL is observable under excitation in the whole transparent region of the sample (including GaAs). Moreover there are no features in PLE spectrum at the GaAs exciton position pointed by the vertical bar in Fig. 2.

3 Analysis

The experimental data presented above can be explained in the following simple model shown schematically in Fig. 3. We suppose that there are a number of deep defect levels around the QDs. The first hint on the existence of the deep levels was found by the observation of Franz–Keldysh oscillations in such kind of structures [7]. Laser light produces transitions between the valence band of the InGaP barrier and defect levels and creates holes which fall into the QDs. The electric current supplies the QDs with electrons. Their recombination with photocreated holes produces AS PL.

There are some features in this model. In steady state conditions we should suppose a presence of an optical transition between the deep levels and the conduction band levels of InGaP (shown by the dashed arrow in Fig. 3) to avoid an accumulation of electrons in the deep level centers. In this sence, AS PL is produced by a two step optical excitation and the electric current. But experimental data shows that the intensity of AS PL is roughly proportional to the optical pump power density ranging from 100 to 1000 W/cm^2.

AS PL also depends roughly linearly on the electric current ranging from 50 to 1000 μA/mm^2. Assuming the efficiency of the electron capturing process by QDs to

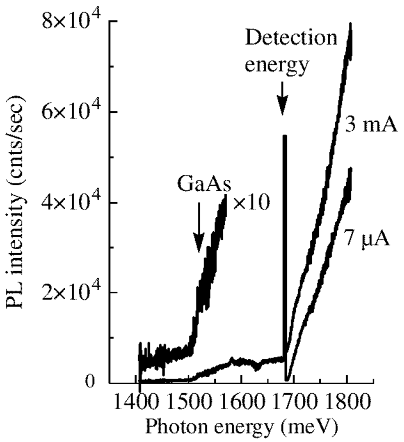

Fig. 2. The PLE spectra of the InP QDs under weak and strong electric currents through the sample. The vertical bar shows the energy position of the GaAs exciton.

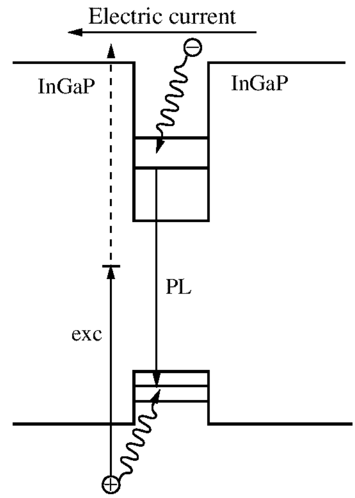

Fig. 3. A simplified model of the processes involved in the observed phenomenon.

be equal to the area of QDs divided by that of the sample we can estimate the number of captured electrons per second per dot to be $n_e = 6 \times 10^6$ for $I = 100\,\mu$A/mm^2. This value of n indicates that the nonradiative channel for electron relaxation is very slow. The nonradiative channel for holes is also slow, because the light excitation intensity of 100 W/cm^2 corresponds to 4×10^6 photons per second per dot. Taking also into account the rather high quantum efficiency of the PL from QDs [ ], we can assume that the deep centers are located in the barrier layer, outside the QDs, but close to the QDs layer.

The observation of the AS PL signal in a wide spectral region of excitation (Fig. 3) can be explained by the broad energy distribution of the initial states for optical pumping belonging to the valence band of InGaP. Therefore we cannot say anything about the energy distribution of the deep level states.

Under resonance excitation, a strong increase of the Stokes PL at positive bias is observed. This effect is very similar with the temperature increase of the PL observed by us [ ]. We think that under resonant excitation a considerable fraction of the PL is a resonant PL hidden by the scattered laser line if there is no electric current. Under applied bias, the electrons supplied by electric current accelerate the relaxation processes due to electron-electron collisions. As a result, a resonant PL is transformed in nonresonant one that causes an increasing of the nonresonant PL.

4 Conclusion

The study performed shows that in structures with InP QDs between InGaP barrier layers, there are defects with deep levels which are located around the QDs. Optical transitions from the InGaP valence band states to the deep levels create the holes which are captured by the QDs. The holes recombine with electrons supplied by the electric current. This process is observable as AS PL. The discovered phenomenon shows that the optical property of InP QDs are essentially affected by the defects in the QDs layer even in high quality structures.

The nature of these deep levels is unknown and further study is needed.

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